

TITLE: DE HAAS VAN ALPHEN EFFECT IN HIGH TEMPERATURE SUPERCONDUCTORS

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**MASTER**

# DE HAAS VAN ALPHEN EFFECT in high temperature superconductors

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## Abstract

The de Haas van Alphen effect has been investigated using 100 tesla fields and temperatures below 4.2 Kelvin. It is concluded from these measurements that YBCO exhibits a Fermi surface with at least three separate extremal frequencies for the c-axis parallel to the applied field. Orbital masses, renormalization  $g$ 's, and scattering rates are estimated from an amplitude analysis.

The focus of this Workshop is on the nature of the ground state of the Ceramic Oxide Superconductors.<sup>1</sup> Recently much progress in understanding these materials has been made by means of angular resolved photo-emission spectroscopy.<sup>2</sup> Although these and similar data have been used to suggest<sup>1</sup> that such materials have a metallic ground state, it is arguable that the photo-excitation process itself, and subsequent electron transport to and alteration by the crystalline surface so modify results that these data might not constitute incontrovertible evidence. Similarly, evidence from positron annihilation continues to improve.<sup>3</sup> In understanding difficult materials, the confluence of evidence on a single question plays a more significant role perhaps than results from any single technique.

A direct ground state measurement such as the de Haas-van Alphen (dHvA) effect has been called for a number of times. This has recently been carried out using 100 Tesla fields and temperatures below 4.2 K by Fowler et al.<sup>XX</sup> We review those results here. The basic data obtained is the oscillatory part of the YBCO magnetization as a function of applied field. As noted below, at least three well-defined frequencies have been deduced from these data. We have reported preliminary results,<sup>4</sup> and recently there has been a report of dHvA oscillations for YBCO by Kido et al. using fields up to 27 T.<sup>5</sup> These data will be reviewed by the next speaker. Their independent measurement whose frequency is about 0.54 kT is virtually identical with our lowest and strongest frequency as is discussed below.

Our dHvA experiments are carried out in large pulsed fields, and this puts a number of rather severe constraints on the preparation of the samples as well as the measurement techniques. The samples are in the form of fine-grained powders embedded in a non-conducting medium so that pulsed magnetic fields can easily penetrate between and into the grains during the time of the pulse. The YBCO samples exhibited a  $T_c$  onset of 93 K. The technique used to prepare and characterize the YBCO samples has been discussed.<sup>6</sup> Superconducting powder was well stirred into epoxy and allowed to harden for 12 hours in a 4.2 T field, oriented along the cylindrical axis.<sup>7</sup> This technique produced samples which maintained a c-axis orientation along the magnetic field axis to better than two degrees as seen by x-rays and NMR. The sample density (17% by volume) is well below the percolation threshold (roughly 31%) as confirmed by scanning electron microscopy. We have considered penetration retardation by eddy current effects in some detail.<sup>8</sup> The thermal rise due to these effects in a typical dHvA experiment is estimated to be no bigger than 0.2 K for our YBCO powder.

The magnetization signals were detected by measuring the voltage developed across a multiturn pickup coil wound on the samples. Because these signals were much smaller than the  $dB/dt$  voltage generated by the magnetic field pulse, a similar "empty" coil containing an inert sample was connected in opposition to the sample "full" coil. Each coil consisted of a single layer of about 100 turns of 30 mm-diameter, insulated copper wire. The full sample was machined from epoxy impregnated with YBCO powder while the empty coil was machined from straight epoxy. Both samples were accurately

machined to 1 mm diameter x 4.5 mm long. Test experiments showed that a typical coil pair, with leads, was balanced to within one turn over the duration of the 100 T pulse. Still, even with only one equivalent uncompensated turn, the amplitude of the dB/dt signal from the coil pair could be substantially larger than the magnetization signals. However, the frequency spectrum of dB/dt itself has little content over 100 kHz and virtually none above 500 kHz. The output of the coil pair was, therefore, fed through a two-stage high band-pass filter that passed only those frequency components above 500 kHz into a 50  $\Omega$  terminated load. The use of carefully balanced coils, together with the passive filter eliminated most signals arising from dB/dt of the field pulse. At the same time, however, subsequent analysis of the true magnetization signals showed that they were very little affected by the filter.

The 100 T fields were obtained from explosive-driven flux compression generators.<sup>9</sup> Experimental volumes are unusually large for 100 T fields. For these experiments, the field coil is made by machining a 16 mm diameter hole in brass bar-stock, 73 mm long. The magnetic fields are monitored by measuring dB/dt developed in a separate coil, of known area. Field measurements are accurate to within 1-2%. Computer simulations of this flux compressor system show<sup>10</sup> that the fields produced have homogeneity of about three parts in 10<sup>4</sup>, for all times, over the region occupied by the sample and empty coil volume in the central portion of the brass load block. At a nominal 100 T peak field the inhomogeneity would be about 0.03 T. Estimating the effect of such an inhomogeneity using

the band structure shape and masses (discussed below), and a nominal 1 kT Fermi surface cross section, the dHvA amplitude is diminished by about a factor of two. Thus, this system has the high fields, low temperatures, easy flux penetration, high-field homogeneity, and oriented, high quality samples necessary as prerequisites for dHvA measurements.

Electrical signals were monitored by a battery of Tektronix 485 analog scopes and several Tektronix 602 digital scopes. The  $dm/dt$  signals developed across the 50: load such as those used to obtain the solid curve in Fig. 1, were recorded on a number of digital channels run at 2 ns sample rates for somewhat over 20 ms per channel (10240 points per channel).

The entire dHvA system was tested in separate 4 K experiments using 1 mm diameter single-crystalline, unoriented copper powders mixed into BN insulating 0.5 mm powder. The copper powder was about 20% by volume. These separate experiments taken in fields larger than 80 T showed a main dHvA frequency peak of 2.1 kT and amplitudes (63 mV) within 30% of Lifshitz-Kosevich (LK)<sup>11</sup> simulations as we have described in Ref. 8. The  $(1/B)$  Fourier transform of these data described below are presented as an inset to Fig. 2. The agreement of this frequency with the well-known copper neck frequency, 2.2 kT, lends confidence to the technique employed here..

In Fig. 1 we present typical YBCO data as the solid line for a temperature of 2.8 K plotted as a function of  $1/B$ , where  $B$  is the magnetic induction. An examination of the data suggests that oscillations of several spectral features exist on a scale of tens

of mV. To probe spectral content, we have taken a Fourier transform. This is defined as

$$A(F) = \int e^{ipF} V(p) dp , \quad (1)$$

where  $V(p)$  is the detected voltage of the pick-up coil of Fig. 1,  $F$  is a possible dHVA frequency in kT,  $p$  is  $1000/B$ , and  $B(t)$  is the magnetic field in T at the implicit time point  $t$ . We plot the spectral density dissipated in the 50: load in Fig. 2

$$P(F) = |A(F)|^2 , \quad (2)$$

as the shaded area. The integral under this curve is the energy dissipated during the experiment in joules. Note that although the spectral shape in the YBCO and copper experiments are very different, the integrated densities are rather similar. Cooling the YBCO system makes the signal voltages larger, following typical dHVA experience. Our results have a vertical precision of about 2%. One spurious feature that appears to some degree in all our experiments is the structure centered about  $p = 10.8$  on Fig. 1. Tests have shown that this structure is due to the mechanics of the flux compressor itself. The amplitude of the structure varies with the degree of coil pair imbalance, but only lasts for about 1 ms. In our Fourier transforms and wave shape analysis, we have simply treated this structure as "data."

It is clear from an examination of Fig. 2 that the several separate features seen in the shaded spectral density may be harmonically related.

The spectral density of several different YBCO dHvA measurements, as well as the one shown in Fig. 2, suggested that there were 3 independent base frequencies: 0.53, 0.78, and about 3.51 kT, together with several harmonics. We have used these base frequencies and have in addition varied 3 other LK parameters for each frequency:  $m^*$  the orbital mass, TD the Dingle temperature or scattering rate, and  $\phi$  the phase. Only a single amplitude constant was permitted for all frequencies since the same coil calibration applies to all of the frequencies and since the amplitude of the harmonics are determined from that of each fundamental. We have implicitly assumed therefore that the curvature factor for each separate sheet is the same. We conclude that 3 independent frequencies and their LK harmonics are well supported by these data.

The parameters derived from different YBCO dHvA experiments and temperatures were similar. After cycling, the "warmest" 4 K data had about 3 times the rms fitting error of the 2.8 K data and a worse signal-to-noise ratio. Nevertheless the same 3 independent frequencies emerged. The present use of LK wave shape analysis is similar in spirit to that pioneered by Higgins and Lowndes.<sup>12</sup>

A number of theoretical models which allow for a Fermi surface in HTSC have been proposed and reviewed.<sup>1</sup> Here we consider the relationship of band models and local density functional theory to these data. To a great degree all HTSC band calculations are in excellent agreement on a scale of a few mRyd. In Table I, we



present our results and the results of Fermi surface predictions from Argonne and Northwestern<sup>13</sup> and from the Naval Research Laboratory, Carnegie, and William & Mary.<sup>14</sup>

If we compare our results with the theoretical band structure cross sections, a considerable agreement emerges. Since the open small sheet cylinders are oriented along the c or z axis, the crystallographic group allows at least two extremal cross-sections per sheet. (One in the +-X-S-Y plane and one in the T-R-U-Z plane.<sup>13,14</sup>) We compare in Table I the ratio of experimental to theoretical masses in order to extract renormalization g's from the relation  $|m^*| = (1+g)|m_{\text{band}}|$ . As is well known, dHvA masses are harder to extract than extremal cross-sections and hence have less accuracy. The electronic cyclotron masses found are strongly renormalized, but not to an extraordinary degree. The renormalization g's are shown in the range of 1-4. Although some of these g's are comparable with those of A-15 materials, the larger g's are consistent with the higher superconducting critical temperature of YBCO using Nambu-Eliashberg theory.<sup>15</sup> We have not yet observed the larger barrel orbits predicted by the band calculations of both theoretical groups. We plan to use a 200 T system to search for these higher cyclotron frequencies.

The scattering times  $\tau$  are derived from the Dingle temperatures  $T_D$ . In making this comparison we have implicitly assumed that all of  $T_D$  is due to impurities or dislocations. Some of  $T_D$  may be due to field inhomogeneity and phase smearing effects. Nevertheless the evidence is that electrons will be able to complete one orbit before scattering ( $lct \gg 1$ ). This internally consistent independent

evidence demonstrates that a dHvA effect should be visible. Note that the scattering time of the neck is about twice as large as that of the pill and that this orbit derived from the 1-d copper chain structures is more sensitive to the formation of twins, which is consistent with the higher TD in Table I.16 We plan to make a similar analysis on the barrel sheets when measured to make intercomparisons.

We conclude that YBCO has a Fermi surface consistent with LDA predictions, that the renormalization  $g$ 's are consistent with Nambu-Eliashberg predictions of  $T_c$ , and that the internal evidence of the scattering rates and cyclotron frequencies allow the dHvA orbits to be observed. At this Workshop , the Fermi Surface of High Temperature Superconductors has been seen in Photemmission , Positron Annihilation and twice now in dHvA experiments . Ceramic superconductors have Fermi Surfaces.

#### Acknowledgements

Many colleagues have contributed greatly to the success of these measurements over the last two years. I am especially grateful to the co-authors of reference xxx . This work was performed under the auspices of the U. S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences. This work was performed using the Pulsed Magnetic Field Facility of the National High Magnetic Field Laboratory at Los Alamos National Laboratory.

## Figure Captions

- 1) Typical YBCO  $dM/dt$  dHvA pick-up coil signal as a function of  $1000/B$ . These data are for a 2.8 K experiment. As discussed in the text the dashed line has been derived from 3 LK frequencies. These are shown separately displaced for the 0.53, 0.78 and 3.51 kT orbital frequencies as dash-dot, long dash, and dash-dot-dot, respectively.
- 2) The  $(1/B)$  Fourier transform of the pick-up coil signal. Peaks in the power spectral density as a function of  $(1/B)$  probe possible cyclotron extremal cross-sectional frequencies of the dHvA effect. As discussed in the text and in Table I extremals of 0.53, 0.78 and 3.51 kT have been extracted from a LK amplitude analysis. The inset shows data on un-oriented 1 mm copper powder taken in fields above 80 T. The neck frequency of Cu is 2.1 kT.

Table I. Y123 Experimental and Theoretical Fermi Surface Areas and Masses (LANL is this work , ANL-NW and NRL-WM are theoretical band structure results from reference a) and b) respectively, c) the cyclotron frequencies  $\omega_c$  are for a 100 Tesla field and are in terahertz, the scattering times  $\tau_s$  in pico-seconds.

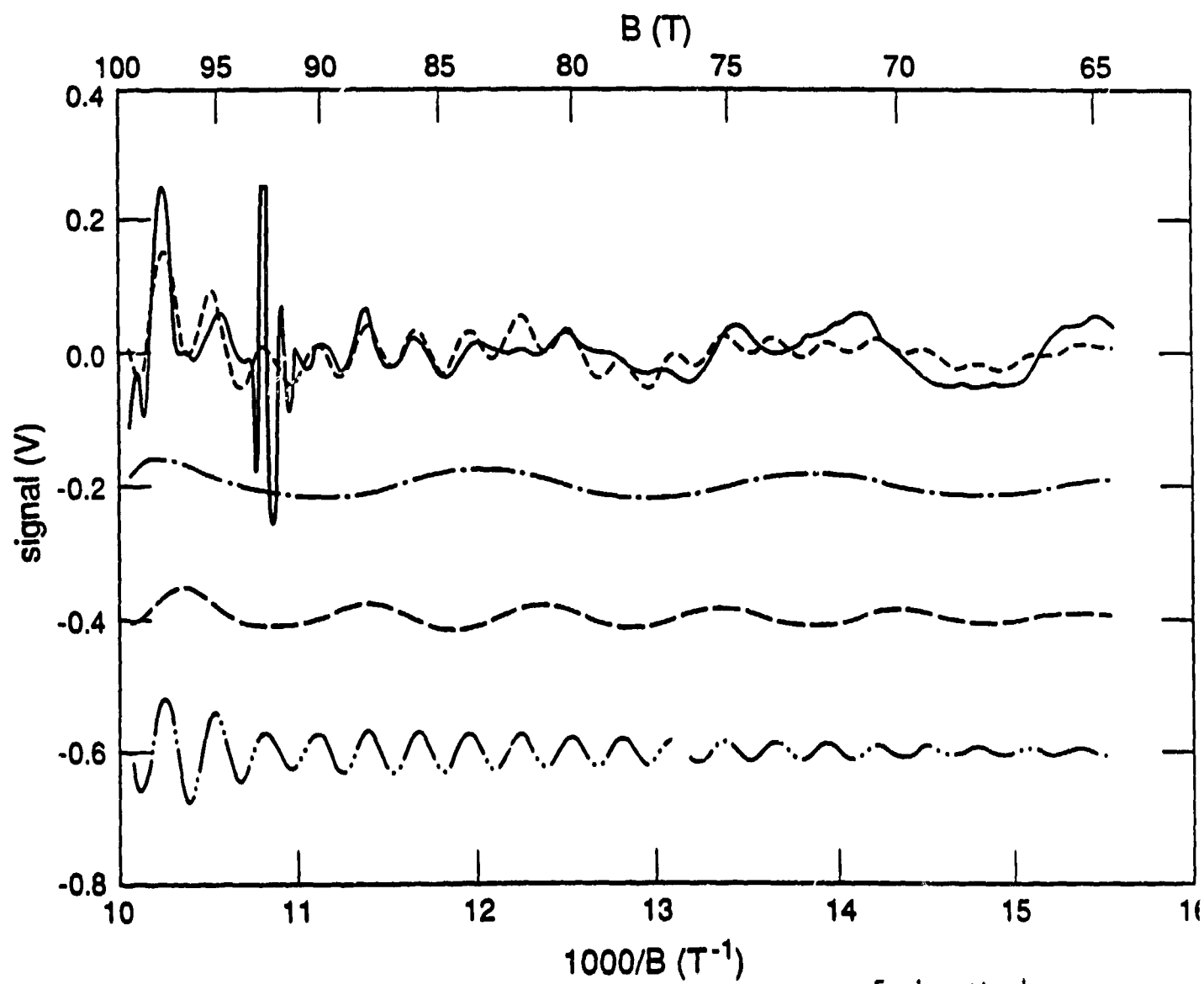
Piece	Area (kT)	Masses (me)	Dingle (K)	Shape	$\omega_c$ (TH)	$\tau_s$ (ps)
Full BZ	27.41					
1) Pill (small cylinder at $k_z = 0$ )						
LANL	0.53+-0.02	7.0+-2.5	1.7+-0.6	(min)	2.5	0.7
ANL-NW	0.40	-2.1				
	1.24	-3.2				
NRL-WM	0.17	-1.6				
2) Pill ( small cylinder at $k_z = \pi/c$ )						
LANL	0.78+-0.02	7.2+-2.5	2.1+-0.7	(max)	2.4	0.6
ANL-NW	0.42	-2.3				
	1.25	-3.8				
NRL-WM	0.22	-1.7				
3) Neck ( 1-d Cu chain in $k_z = \pi/c$ )						
LANL	3.51+-0.10	7.4+-2.6	3.4+-1.2	(min)	2.4	0.4
NRL-WM	3.70	1.45				
4) Barrel (1) (Cu-plane)						
LANL	unobserved					
ANL-NW	12.05	-1.5				
	12.70	-1.3				
NRL-WM	12.41	-1.4				
5) Barrel (2) (Cu-plane)						
LANL	unobserved					
ANL-NW	12.68	-1.6				
	13.84	-1.9				
NRL-WM	12.57	-1.5				

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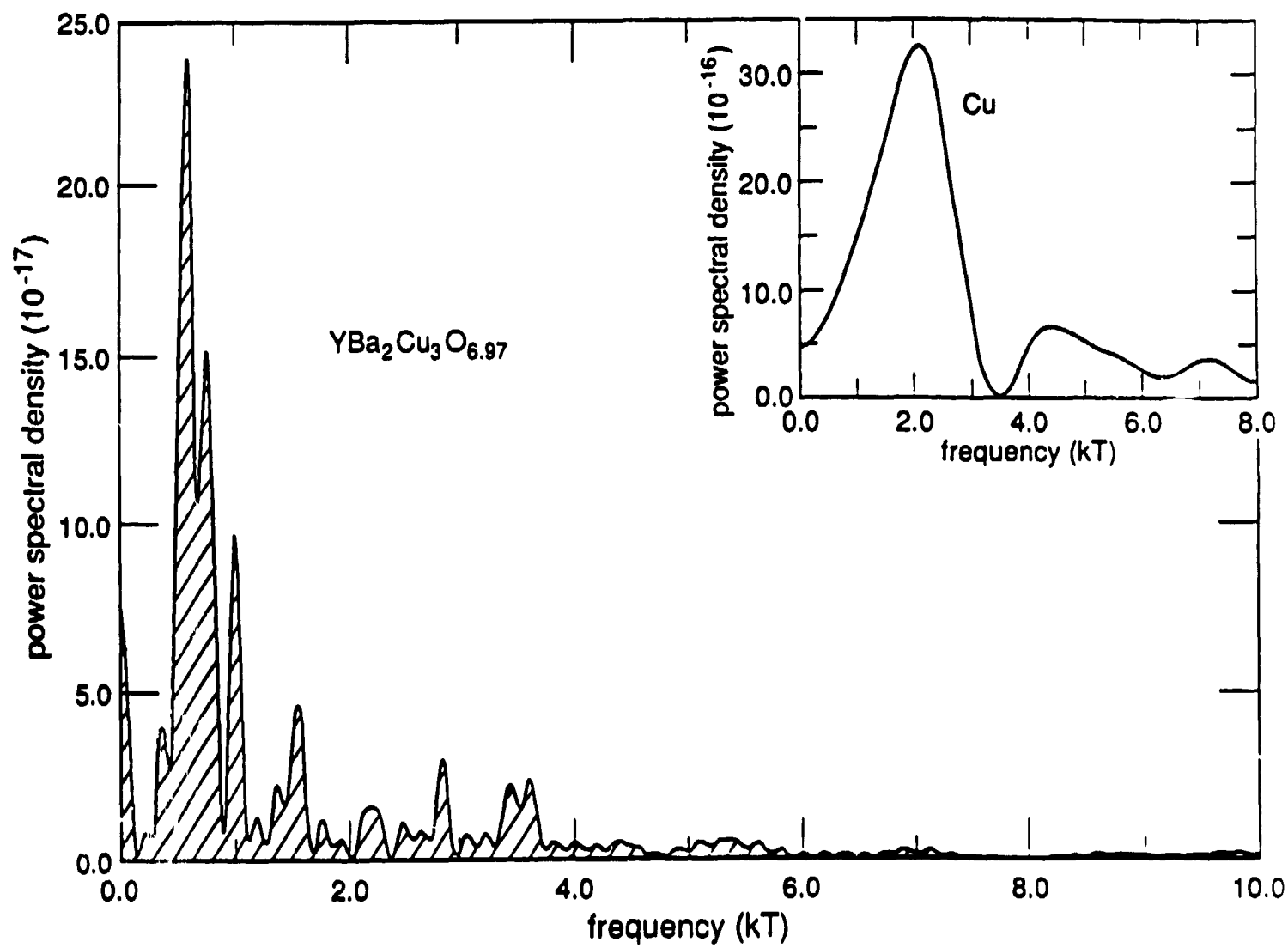
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Fowler et al.

Figure 1





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Figure 2